

FORM PTO-1390 (REV. 11-2000)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER <b>514413-3911</b>	
<b>TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371</b>				U.S. APPLICATION NO. (If known see 37 C.F.R. 1.5) <b>10/049410</b>	
				PRIORITY DATE CLAIMED <b>9 AUGUST 1999</b>	
INTERNATIONAL APPLICATION NO. <b>PCT/EP00/07339</b>		INTERNATIONAL FILING DATE <b>28 JULY 2000</b>			
TITLE OF INVENTION <b>HERBICIDAL COMPOSITIONS COMPRISING POST-EMERGENCE HERBICIDES FOR SOIL APPLICATION</b>					
APPLICANT(S) FOR DO/EO/US <b>Dr. Udo BICKERS and Dr. Gerhard FRISCH</b>					

Applicants herewith submit to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

- ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
- ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
- ☒ This is an express request to promptly begin national examination procedures (35 U.S.C. 371(f)).
- ☒ The US has been elected by the expiration of 19 months from the priority date (PCT Article 31).
- ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  - ☒ is attached hereto (required only if not communicated by the International Bureau).
  - ☐ has been communicated by the International Bureau.
  - ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
- ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
- ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
  - ☐ are attached hereto (required only if not communicated by the International Bureau).
  - ☐ have been communicated by the International Bureau.
  - ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - ☒ have not been made and will not be made.
- ☐ A English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
- ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
- ☐ An English language translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11 to 20 below concern document(s) or information included:

- ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
- ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
- ☒ A FIRST preliminary amendment.
- ☐ A SECOND or SUBSEQUENT preliminary amendment.
- ☐ A substitute specification.
- ☐ A change of power of attorney and/or address letter.
- ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
- ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
- ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
- ☒ Other items or information:  
PCT/RO/101, PCT/IB/308, PCT/ISA/210  
1 page Abstract

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U.S. APPLICATION NO. <b>107049410</b> (37 CFR 1.50)		INTERNATIONAL APPLICATION NO. PCT/EP00/07339		ATTORNEY'S DOCKET NO. 514413-3911	
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21. <input checked="" type="checkbox"/> The following fees are submitted				<b>CALCULATIONS PTO USE ONLY</b>	
<b>BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):</b> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO..... \$1040.00  International preliminary examination fee (37 C.F.R. 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO..... \$890.00  International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO..... \$740.00  International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4)..... \$710.00  International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4)..... \$100.00  <b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				<b>\$ 890.00</b>	
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Total Claims	13 - 20 =	0	x \$18.00	\$ 000.00	
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 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

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SIGNATURE \_\_\_\_\_  
**WILLIAM F. LAWRENCE**  
 NAME \_\_\_\_\_  
 28,029  
 REGISTRATION NUMBER \_\_\_\_\_

Dated: February 7, 2002

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

Applicant: BICKERS, et al.  
Filed.: Filed Concurrently Herewith  
Title of Invention: HERBICIDAL COMPOSITIONS COMPRISING POST-  
EMERGENCE HERBICIDES FOR SOIL APPLICATION

745 Fifth Avenue  
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**PRELIMINARY AMENDMENT**

Assistant Commissioner for Patents  
Box PCT  
Washington, D.C. 20231

Sir:

Before the issuance of the first Office Action, please amend the above-identified application as follows:

**IN THE SPECIFICATION:**

Please replace the paragraph beginning at page 2, line 26, with the following rewritten paragraph:

In the field of crop protection and, accordingly, also in the field of the control of harmful plants, there have been repeated attempts to enhance the use of properties of a given active compound, such as, for example, its activity spectrum, its persistence or the required application rate, for example by modified formulations or application methods. Thus, Weed Research, 1997, 37, 19-36, examines whether the activity of the herbicide chlorsulfuron can be enhanced and its tendency to leach can be reduced by using a controlled-release formulation. US 5,674,519 discloses that the tendency of certain crop protection agents including herbicides to leach in the soil can be reduced by a formulation in which the active compounds are present in microencapsulated form. Similarly, WO 98/05205 and US 5,543,383 describe a particular form of encapsulating crop protection agents which leads to an increased activity and a reduced susceptibility to leaching by rain. Furthermore, WO 99/26474 describes methods for releasing active compounds using cyclodextrins or carbohydrates. EP-A 0 517 669 discloses that the leaching behavior of agrochemicals can be improved by microencapsulation in polyester polymers. GB-A 1 041 028 provides mixtures of paraquat salts and fuller's earth having anticoccidial properties.

Please replace the paragraph beginning at page 3, line 8, with the following rewritten paragraph: Accordingly, it is an object of the present invention to make possible the use of post-emergence herbicides by the pre-emergence method. This object is achieved by herbicidal compositions, comprising an effective amount of one or more post-emergence herbicides and an amount of a carrier material from the group consisting of fullers earth, aerogels, high-molecular-weight polyglycols and polymers based on acrylic acid, methacrylic acid and copolymers thereof, with

the proviso that herbicidal compositions comprising paraquat and fuller's earth shall be excluded.

Please replace the paragraph beginning at page 3, line 27, with the following rewritten paragraph:

Usually, the compositions according to the invention comprise

- a) from 0.15 to 48% by weight of one or more post-emergence herbicides,
- b) from 2 to 90% by weight of a carrier material and
- c) from 0 to 97% by weight of a solvent.

#### **IN THE CLAIMS:**

Please amend claim 1 as follows:

1. (Amended) A herbicidal composition, comprising an effective amount of one or more post-emergence herbicides and an amount of a carrier material selected from the group consisting of fuller's earth, aerogels, high-molecular-weight polyglycols and polymers based on acrylic acid, methacrylic acid and copolymers thereof, with the proviso that herbicidal compositions comprising paraquat and fuller's earth shall be excluded.

#### **REMARKS**

The Specification and Claim 1 has been amended to reflect the amendments made under PCT Article 34. Attached hereto is a marked up version of the changes made by the current amendment. The attached page is captioned "**Version with markings to show changes made.**"

The filing fee has been calculated based upon the amendment made to the claim 1.

Respectfully submitted,

FROMMER LAWRENCE & HAUG LLP  
Attorneys for Applicant

By:   
William F. Lawrence  
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Tel. (212) 588-0800

2022.07.04 10:44:13

**VERSION WITH MARKINGS TO SHOW CHANGES MADE****In the Specification:**

Paragraph beginning at line 26 of page 2 has been amended as follows:

In the field of crop protection and, accordingly, also in the field of the control of harmful plants, there have been repeated attempts to enhance the use of properties of a given active compound, such as, for example, its activity spectrum, its persistence or the required application rate, for example by modified formulations or application methods. Thus, Weed Research, 1997, 37, 19-36, examines whether the activity of the herbicide chlorsulfuron can be enhanced and its tendency to leach can be reduced by using a controlled-release formulation. US 5,674,519 discloses that the tendency of certain crop protection agents including herbicides to leach in the soil can be reduced by a formulation in which the active compounds are present in microencapsulated form. Similarly, WO 98/05205[too,] and US 5,543,383 describe a particular form of encapsulating crop protection agents which leads to an increased activity and a reduced susceptibility to leaching by rain. Furthermore, WO 99/26474 [and WO 99/26743] describes methods for releasing active compounds using cyclodextrins or carbohydrates. EP-A 0 517 669 discloses that the leaching behavior of agrochemicals can be improved by microencapsulation in polyester polymers. GB-A 1 041 028 provides mixtures of paraquat salts and fuller's earth having anticoccidial properties.

Paragraph beginning at line 8 of page 3 has been amended as follows:

Accordingly, it is an object of the present invention to make possible the use of post-emergence herbicides by the pre-emergence method. This object is achieved by herbicidal compositions, comprising an effective amount of one or more post-emergence herbicides and an amount of a

carrier material from the group consisting of fullers earth, aerogels, high-molecular-weight polyglycols and polymers based on acrylic acid, methacrylic acid and copolymers thereof, with the proviso that herbicidal compositions comprising paraquat and fuller's earth shall be excluded.

Paragraph beginning at line 27 of page 3 has been amended as follows:

Usually, the compositions according to the invention comprise

- a) from [0.001] 0.15 to 48% by weight of one or more post-emergence herbicides,
- b) from 2 to 90% by weight of a carrier material and
- c) from 0 to 97% by weight of a solvent.

**In the Claims:**

Claim 1 has been amended as follows:

1. (Amended) A herbicidal composition, comprising an effective amount of one or more post-emergence herbicides and an amount of a carrier material selected from the group consisting of fuller's earth, aerogels, high-molecular-weight polyglycols and polymers based on acrylic acid, methacrylic acid and copolymers thereof, with the proviso that herbicidal compositions comprising paraquat and fuller's earth shall be excluded.

WO 01/10211

PCT/EP00/07339

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## Description

Herbicidal compositions comprising post-emergence herbicides for soil application

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The invention relates to the technical field of herbicides, and in particular of herbicides for soil application.

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For controlling undesirable harmful plants, the user can choose from a large number of herbicides which can be applied depending on the biological properties of the herbicides, the type of harmful plants to be controlled and the type of useful plants. Here, it has also to be taken into account that numerous herbicides, owing to their intrinsic mechanism of action, can be used either exclusively by the pre-emergence method or exclusively by the post-emergence method. Both methods and the herbicides used for these methods have certain advantages and disadvantages. Disadvantages of the post-emergence method where, for example, herbicides with foliar action such as bilanafos, diquat, glufosinate-ammonium, glyphosate and paraquat are also used, are considered to be, for example:

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- damage to useful plants by overdosage
- depending on the type of herbicide used, on the harmful plants to be controlled and/or the useful plants in question, a need to repeat the application several times.

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The last mentioned disadvantage is particularly grave, especially from the point of view of time-economic farming.

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On the other hand, numerous herbicides which can be used by the post-emergence method have considerable advantages, in particular also from an ecological point of view, since in many cases they have more favorable toxicological and ecotoxicological properties than the herbicides which can be used by the pre-emergence method. Depending on the particular application situation, in many cases it would therefore be desirable for the user to be able to use a post-emergence herbicide under the conditions of a pre-emergence application, i.e. prior to the emergence of the plants or, if appropriate, simultaneously with the sowing of the useful plants. However, hitherto such an application has, in particular in the case of herbicides with foliar action, not been possible, for reasons of the mechanism of action on

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which these herbicides are based, since herbicides with foliar action in the form of the herbicidal compositions of the prior art, for example, are only taken up via the green parts of the plant, for reasons of their leaching properties or else for reasons of their degradation behavior in the soil.

- 5 Glufosinate-ammonium (2-amino-4-(hydroxymethylphosphinyl)butanoic acid), for example, which can be used as herbicide with foliar action is known to be decomposed rapidly in the soil, so that it is not capable of displaying any herbicidal action (G. Hoerlein in "Reviews of Environmental Contamination and Toxicology", vol. 138, Springer-Verlag; "The Pesticide
- 10 Manual", 11th Edition, 1997, British Crop Protection Council). The herbicide glyphosate (N-(phosphonomethyl)glycine), which likewise has foliar action, is known to be adsorbed strongly by the soil and to be degraded therein, so that it is not available to the plant in sufficient amounts (L. Torstensson in "The Herbicide Glyphosate", Butterworths, pp. 137-150).

- 15 Hereinbelow, the term "herbicide" is, depending on the context, to be understood as meaning the pure herbicidally active compound or the herbicidally active compound in formulated form, i.e. the herbicidal composition. The terms "pre-emergence" and "post-emergence" are here to
- 20 be understood as referring to the time of the emergence of the harmful plants, i.e. a treatment by the pre-emergence method means a treatment with a herbicide prior to the emergence of the harmful plants and thus prior to the formation of green parts of plants. Analogously, a treatment by the post-emergence method means a treatment with a herbicide after
- 25 emergence of the harmful plants.

- In the field of crop protection and, accordingly, also in the field of the control of harmful plants, there have been repeated attempts to enhance the use properties of a given active compound, such as, for example, its
- 30 activity spectrum, its persistence or the required application rate, for example by modified formulations or application methods. Thus, Weed Research, 1997, 37, 19-26 examines whether the activity of the herbicide chlorsulfuron can be enhanced and its tendency to leach can be reduced by using a controlled-release formulation. US 5,674,519 discloses that the
- 35 tendency of certain crop protection agents including herbicides to leach in the soil can be reduced by a formulation in which the active compounds are present in microencapsulated form. WO 98/05205, too, describes a particular form of encapsulating crop protection agents which leads to an increased activity and a reduced susceptibility to leaching by rain.

Furthermore, WO 99/26474 and WO 99/26743 describe methods for releasing active compounds using cyclodextrins or carbohydrates.

- 5 However, none of the publications mentioned indicates that it may be possible to use post-emergence herbicides pre-emergence by employing suitable measures.

10 Accordingly, it is an object of the present invention to make possible the use of post-emergence herbicides by the pre-emergence method. This object is achieved by herbicidal compositions, comprising an effective amount of one or more post-emergence herbicides and an amount of a carrier material from the group consisting of fullers earth, aerogels, high-molecular-weight polyglycols and polymers based on acrylic acid, methacrylic acid and copolymers thereof.

15 Here, the compositions according to the invention should only comprise post-emergence herbicides which, in the form of the herbicidal compositions of the prior art, can be used exclusively by the post-emergence method, i.e. after emergence of the undesirable harmful plants.

20 Surprisingly, post-emergence herbicides which are present bound to the carrier materials mentioned above display herbicidal action against undesirable harmful plants when used by the pre-emergence method, i.e. when the herbicidal compositions have been applied prior to the  
25 emergence of the harmful plants.

Usually, the compositions according to the invention comprise

- a) from 0.001 to 48% by weight of one or more post-emergence herbicides,  
30 b) from 2 to 90% by weight of a carrier material and  
c) from 0 to 97% by weight of a solvent.

Particularly suitable post-emergence herbicides are those from the group of the herbicides with foliar action. Preference is given to herbicides from the  
35 group consisting of bilanafos, diquat, glufosinate-ammonium, glyphosate and paraquat. Particular preference is given to glufosinate-ammonium. The abovementioned herbicides are known, for example, from "The Pesticide Manual", 11th Edition, 1997, British Crop Protection Council. The herbicides in question can, of course, also be employed in the commercial

salt form, as known, for example, from "The Pesticide Manual".

- It is a further advantage of the compositions according to the invention that they are present in solid form and can be used, for example, in the form of granules. The user can apply this solid presentation form directly to the area to be treated, i.e. without it being necessary to prepare a spray liquor. To this end, they are, depending on the type of useful plants and the type of expected harmful plants to be controlled, applied onto the soil to be treated, worked into the soil or applied by side application.
- Side application means that the herbicide is applied in the soil below the seeds. Here, it is particularly advantageous that the application of the herbicidal compositions can also take place in the same operation as the sowing of the crop plants, resulting in the soil being subjected to reduced mechanical stress.
- The herbicides which can be used for the compositions according to the invention can be processed to the compositions according to the invention in the form of the pure active compound or else in the customary commercial formulation together with carrier materials and, if appropriate, additional substances, in the manner described further below. A particular advantage of the compositions according to the invention is their good environmental compatibility. Owing to the fact that the herbicides, in contrast to conventional application methods, are not sprayed in liquid form but applied in solid form, there is no risk of uncontrolled drift of spray mists onto the user and adjacent areas and plants. Moreover, environmentally compatible formulation auxiliaries are used as they are either - like the high-molecular-weight polyglycols and polymers of acrylic acid, methacrylic acid and copolymers thereof - biodegradable to unobjectionable substances or - like fuller's earth - show environmentally neutral behavior.
- The herbicidal compositions are prepared by mixing the active compounds, carrier materials and, if appropriate, further additives in question according to methods known to the person skilled in the art. Granules, for example, can be prepared by the methods described in EP-A - 0 413 267. Depending on the carrier material, the substances in question can be present in solid or liquid or dissolved form. In the preparation of granules, it is advantageous for the active compounds and additives to be incorporated to be present in liquid or dissolved form. Here, these substances can be applied uniformly in the known manner for example by spraying, pouring,

dropwise addition, treatment in a fluidized bed, concrete mixer, tumble mixer, etc. It is also possible to oversaturate these carrier materials with solutions of the active compounds and additives in question to form, for example, gels, or to remove excess water by drying. In the case of the preparation of melt granules, for example melt granules based on polyethylene glycol, active compounds and additives are preferably incorporated in solid form and extruded. Also possible is tableting, pelletizing, the preparation of flakes and comminution to powders by breaking or grinding. In all preparation processes, aftertreatment by comminution may take place. It is also possible to compact very fine granules using suitable adhesives to avoid the formation of dust. It is furthermore possible, if appropriate, to add additives to improve, for example, the flowability of the granules or their wettability.

- 15 Depending on the intended purpose, the herbicidal compositions may additionally comprise other substances which can be used in crop protection, such as pre-emergence herbicides, plant growth regulators, fungicides, insecticides, safeners ((herbicidal) antidotes), nutrients, preservatives, seed dressings and fertilizers. The selection of the pre-emergence herbicides, plant growth regulators, fungicides, insecticides and seed dressings to be added, if appropriate, depends on the type of useful plants and the type of expected harmful plants, fungi, insects and feed pests to be controlled. In principle, it is possible to use all commercial substances which are usually employed in crop protection. Suitable nutrients and fertilizers are, in particular, aqueous ammonium nitrate/urea solutions and NPK solutions such as 12-6-8, 8-8-6, 5-8-10, and also ammonium sulfate and/or ammonium nitrate solutions.

- 30 Preferred carrier materials are aerogels, polymers based on acrylic acid, methacrylic acid and copolymers thereof and also high-molecular-weight polyglycols.

- When using the compositions according to the invention, it has to be ensured that the useful plant is sufficiently tolerant to the herbicide employed. In addition to useful plants in which sufficient tolerance to herbicides has been achieved by targeted genetic manipulation, this includes plants which, such as, for example, barley, have sufficient tolerance to some herbicides even without genetic manipulations. The compositions according to the invention can be used particularly

advantageously in crops of useful plants from the group consisting of cereals, corn, soyabean and rapeseed. They are preferably used in crops which are sufficiently tolerant to the herbicide in question owing to genetic manipulation.

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As in the case of other herbicidal compositions, it is, of course, in the case of the compositions according to the invention also possible and in some cases advantageous to provide the herbicide with additives such as surfactants, wetting agents, emulsifiers, adjuvants, ammonium salts, 10 preservatives, colorants, antifoams, tackifiers, solvents, buffer systems and UV stabilizers. Depending on the type and purpose of the additives, these can be processed together with the herbicide in a formulation or else be present and subsequently used separately from the herbicide. These additives serve to improve application properties. The use of such additives 15 may be advantageous to enhance, for example, the persistence of the herbicide. Preservatives are used, for example, to slow down the biological degradation of active compounds and/or formulation auxiliaries.

Suitable surfactants are, for example, condensates of formaldehyde and 20 phenol and/or naphthol, in each case with or without sodium bisulfite, such as Tamol NNO from BASF, Rapidamin-Reserve C from Clariant or Galoryl MT800 or DT201 from CFPI; C<sub>12</sub>-C<sub>24</sub> fatty alcohols having 2 to 40 EO and/or PO, if appropriate phosphated and/or neutralized with alkanolamine, alkali metal or ammonia; di- and tristyrylphenyl analogs of the fatty alcohols 25 mentioned above, such as, for example, the Genapol series from Clariant, Grafol types from Henkel and Soprophor types from Rhodia; alkyl ether sulfates such as Genapol LRO® from Clariant; alkyl- and/or alkenylsulfonates such as Hostapur OS from Clariant, ligninsulfonates such as Ufoxane 3A and Vanisperse CB from Booregard; Reax types from 30 Westvaco; N-methyltaurides such as Hostapon T from Clariant; sulfosuccinic monoester salt such as Hoe S 1728 from Clariant; alkyl polysaccharides such as Plantaren APG 600 from Henkel; ethoxylated C<sub>12</sub>-C<sub>24</sub>-fatty amines, such as the Genamin types from Clariant. Suitable solvents are, in each case depending on the intended purpose, alcohols, 35 diols, polyols, N-substituted pyrrolidones, ketones, aldehydes, ethers, polyethers, paraffins, aromatic compounds, heteroaromatic compounds, cycloalkanones, dimethylsulfoxide, tetrahydrofuran and water. It is advantageous to employ water.

Suitable preservatives are, for example, Bronidox L from Henkel, Mergal types from Riedel de Haen, Proxel from ICI, ascorbic acid derivatives, benzoic acid derivatives, formaldehyde, citric acid, preservatives from the Kathon series from Rohm & Haas and brohopol from BASF.

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Suitable adjuvants are, for example, alkyl polysaccharides or lauryl ether sulfates.

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Suitable carrier materials from the group of the high-molecular-weight polyglycols are, in particular, polyethylene glycols having a molecular weight of from 2000 to 40,000 (PEG 2000 to PEG 40,000). Suitable carrier materials from the group of the aerogels are, in particular, the aerogels described in EP-A 0 171 722.

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It is particularly advantageous to add a substance such as ammonium sulfate, ammonium nitrate and one of the surfactants mentioned above, in particular a surfactant from the group of the alkyl ether sulfates.

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The use of the herbicidal compositions by the pre-emergence method is novel and also forms part of the subject matter of the invention.

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Using the herbicidal compositions according to the invention, it is possible to control undesirable harmful plants by the pre-emergence method. This method is new and also forms part of the subject matter of the invention.

The invention is illustrated by the embodiments that follow.

#### **A. Formulation examples**

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By way of example, the qualitative and quantitative (in % by weight) composition of numerous herbicidal compositions according to the invention is listed in Tables 1 to 4.

Table 1

No.	Herbicide (active compound)		Carrier materials			Aerogel		Aerogel		Additives	
	Basta 50% (glu- fosinate)	Basta 150 g/l (glu- fosinate)	Lugusorb AF1	Lugusorb AF2	P88	C373	Water				
1	2			90			8.00				
2	2			10			88.00				
3	2			4			94.00				
4	2			2			96.00				
5	2		90				8.00				
6	2		88				10.00				
7	2		50				48.00				
8	2		10				88.00				
9	2		4				94.00				
10	2		2				96.00				
11		95	5								
12		90	10								
13	90		10								
14	95		5								
15	10		90								
16	2										
17	2				90		8.00				
18	2				80		18.00				
19	2				40		58.00				
20	2				50		48.00				
21	2				65		33.00				
22	2					60	38.00				
						50	48.00				

Basta 150 g/l: 13.5% of glufosinate-ammonium, 58.81% of Genapol LRO, 10.0% of Dowanol PM, 0.25% of Fluowet PL80, 0.005% of Duasyn acid blue AE, water ad 100%

Table 2

Herbicide		Carrier materials			Additives														
n.o.	@Basta 50%	@Basta 99%	Luqasorb AF1	Aerogel P88	Aerogel C373	ammonium nitrate	ammonium sulfate	urea	Hostapur OSB	Geropon SDS	Genapol LRO	Sipemat 22S	Duasyn acid blue AF	Silicolapse 5020	Stepanol ME Dry	propylene glycol	rapeseed oil	glycerol	
23	2				40														
24	2				35														
25	2				30														
26	85	15																	
27	95	5																	
28	2		81			1			5			5	0.01			5.99			
29	2		81			1						5	0.01		5	5.99			
30	2		86					1				5	0.01			5.99			
31	2		86					5				5	0.01			1.99			
32	2			40						10	10		0.05	4		42.95			
33	2		40					1		10	10		0.05	4		39.25		10	
34	85		5								10								
35	85		5																
36	85		5																
37	11		84														10		
38	80		5									5							
39	80		5															6	
40	2		75																
41	2		75			1						5							
42	2		75			4						5							
43	2		75				1					5							
44		1				4						5							
45		1														99			
						1										98			





Table 1a - Combinations

	Herbicide						Carrier material		Additives								
no.	Basta 50%	Basta 99%	mono-linuron tech. grade	CMP tech. Grade	simazine tech. grade	diuron tech. Grade	dichloro- prop-p-DMA tech. grade	2,4-d-DMA salt	Aerogel P88	polyethylene glycol 20,000	propylene glycol	Hostapur OSB	ammonium nitrate	Bronidox L	duasyn acid blue AE	Stepanol ME dry	Megal K9N
87	2.0								40.0		46.85		1.0		0.05	10.0	0.1
88			1.1						40.0		47.80		1.00		0.05	10.0	0.1
89			1.05						40.0		47.85	10.0	1.00	0.05	0.05		
90			1.05							98.95							
91		0.1	1.05							98.85							
92		0.3	1.05							98.65							
93		0.5	1.05							98.45							
94		1.0	0.50							98.50							
95		1.0	1.05							97.95							
96		1.0	2.10							96.90							
97				1.2					40.0		47.25	10.0	1.0	0.5	0.05		
98	2.0			0.3					40.0		46.55		1.0		0.05	10.0	0.1
99	2.0			0.1					40.0		46.75		1.0		0.05	10.0	0.1
100	2.0				1.2	1.9			40.0		43.79		1.0		0.01	10.0	0.1
101						1.5			40.0		47.39		1.0		0.01	10.0	0.1
102	2.0					4.0			40.0		42.89		1.0		0.01	10.0	0.1
103					3.0				40.0		45.89		1.0		0.01	10.0	0.1
104	2.0				3.0				40.0		43.89		1.0		0.01	10.0	0.1
105							1.2		40.0		47.89		1.0		0.01	10.0	0.1
106	2.0						0.2		40.0		46.69		1.0		0.01	10.0	0.1
107	2.0						0.3		40.0		46.59		1.0		0.01	10.0	0.1
108	2.0						0.5		40.0		46.39		1.0		0.01	10.0	0.1
109											47.74		1.0		0.01	10.0	0.05
110	2.0							1.2	40.0		46.44		1.0		0.01	10.0	0.05
								0.5	40.0				1.0		0.01	10.0	0.05



Table 3a

	Herbicide	Carrier material	Additives											
no.	Basta 50%	Aerogel P88	propylene glycol	Hostapur	ammonium nitrate	Bronidox L	duasyn acid blue AE	Geropon	Stepanol	Mergal K9N	phenyl-succinic acid	1-naphthyl-acetic acid Na	3-indolyl-acetic acid Na	glibberelic acid Na
134	2.0	40.0	46.35		1.0		0.5		10.0	0.1	0.5			
135	2.0	40.0	44.85		1.0		0.05		10.0	0.1	2.0			
136	2.0	40.0	46.75		1.0		0.05		10.0	0.1		0.1		
137	2.0	40.0	45.85		1.0		0.05		10.0	0.1		1.0		
138	2.0	40.0	46.44	10.0	1.0	0.5	0.05						0.1	
139	2.0	40.0	46.25	10.0	1.0	0.5	0.05						0.20	
140	2.0	40.0	46.84		1.0		0.05		10.0	0.1				0.01
141	2.0	40.0	46.65		1.0		0.05		10.0	0.1				0.2

## B Biological examples

Meanings of the abbreviations used below:

5	ABUTH	Abutilon theophrasti	AMARE	Amaranthus retroflexus
	BRSNW	Brassica napus	CHEAL	Chenopodium album
	GALAP	Galium aparine	HORVS	Hordeum vulgare
	LOLMU	Lolium multiflorum		

### 10 B.1 Action against harmful plants of glufosinate-ammonium bound to a polymer carrier material, compared to that of glufosinate-ammonium applied by a conventional method

In a greenhouse, a granulous formulation, prepared according to example 48, of the herbicide glufosinate-ammonium was broadcast at an application rate of 500 g of active compound per hectare (converted) on humus-containing loam soil, and the soil was then watered. Three days after the application, seeds of *Setaria viridis* were sown at a depth of 1 cm. For the entire duration of the experiment, a daytime temperature of 22-24°C and a nighttime temperature of 16-18°C were maintained, a uniform day length of 16 hours being achieved by additional illumination with sodium vapor lamps (approximately 7000lux). The relative atmospheric humidity was 60-80%. Visual scoring 35 days after sowing showed that an activity of 70% against *Setaria viridis* was achieved. For comparison, glufosinate-ammonium was applied as an aqueous solution, under otherwise identical conditions and likewise at an application rate of 500 g of active compound per hectare (converted). Visual scoring 35 days after sowing showed that the activity achieved against *Setaria viridis* was nil (0%).

### 30 B.2 Comparison of the action against harmful and useful plants of glufosinate-ammonium bound to polymer carrier material

In a greenhouse, seeds of the harmful plants LOLMU, AMARE and CHEAL and the useful plant HORVS were sown on humus-containing loam soil, covered with a 0.5 cm layer of the same soil and then watered. One day after sowing, a formulation, prepared according to example 48, of the herbicide glufosinate-ammonium was broadcast on the soil at an application rate of 1000 g of active compound per hectare (converted). For the entire duration of the experiment, a daytime temperature of 22-24°C and a nighttime temperature of 16-18°C were maintained, a uniform day length of 16 hours being achieved by additional illumination with sodium

vapor lamps (approximately 7000 lux). The relative atmospheric humidity was 60-80%. Visual scoring 35 days after sowing gave the results shown in Table 5, and it was found that, in contrast to the three harmful plants tested, the crop plant HORVS had not been damaged by the herbicide.

5

Table 5 Activity [%] against harmful and useful plants

Treatment with herbicide according to formulation example No.	LOLMU	AMARE	CHEAL	HORVS
48	40	70	60	0

### B.3 Action against harmful plants of glufosinate-ammonium bound to different carrier materials

- 10 Seeds of the harmful plants GALAP, AMARE, CHEAL and LOLMU were sown outdoors on humus-containing loam soil, covered with a 0.5 cm layer of the same soil and then watered. One day after sowing, formulations, prepared according to examples No. 40, 45 and 46, of the herbicide glufosinate-ammonium were broadcast at an application rate of 750 g of
- 15 active compound per hectare (converted) on the soil. Visual scoring 35 days after sowing gave the results shown in Table 6.

Table 6 Activity [%] against harmful plants

Treatment with herbicide according to formulation example No.	GALAP	AMARE	CHEAL	LOLMU
40	80	10	15	25
45	100	35	70	30
56	70	40	85	25

### 20 B.4 Effect of different fertilizers on the efficacy of glufosinate-ammonium bound to polymer carrier material

- In a greenhouse, seeds of the harmful plants ABUTH, AMARE, CHEAL and LOLMU were sown in humus-containing loam soil, covered with a 0.5 cm layer of the same soil and then watered. One day after sowing, a
- 25 formulation, prepared according to examples 41 and 43, of the herbicide glufosinate-ammonium was broadcast at an application rate of 750 g of active compound per hectare (converted) on the soil. For the entire duration of the experiment, a daytime temperature of 22-24°C and a nighttime temperature of 16-18°C were maintained, a uniform day length of

16 hours being achieved by additional illumination with sodium vapor lamps (about 7000 lux). The relative atmospheric humidity was 60-80%. Visual scoring 35 days after sowing gave the results shown in Table 7.

5 Table 7 Activity [%] against harmful plants

Treatment with herbicide according to formulation example No.	ABUTH	AMARE	CHEAL	LOLMU
41	80	90	85	15
43	75	75	75	10

#### B.5 Effect of different surfactants on the efficacy of glufosinate-ammonium bound to carrier materials

- 10 In a greenhouse, seeds of the harmful plants GALAP, AMARE, CHEAL and LOLMU were sown in humus-containing loam soil, covered with a 0.5 cm layer of the same soil and then watered. One day after sowing, a formulation, prepared according to examples 40, 29 and 28, of the herbicide glufosinate-ammonium was broadcast at an application rate of 750 g of active compound per hectare (converted) on the soil. For the
- 15 entire duration of the experiment, a daytime temperature of 22-24°C and a nighttime temperature of 16-18°C were maintained, a uniform day length of 16 hours being achieved by additional illumination with sodium vapor lamps (about 7000 lux). The relative atmospheric humidity was 60-80%. Visual scoring 35 days after sowing gave the results which showed that the
- 20 efficacy of the herbicide is enhanced by adding surfactants to glufosinate-ammonium bound to carrier materials, see Table 8.

Table 8 Activity [%] against harmful plants

Treatment with herbicide according to formulation example No.	GALAP	AMARE	CHEAL	LOLMU
40 (without surfactant)	80	10	15	25
28 (with surfactant)	98	60	98	60
29 (with surfactant)	90	60	90	40

#### 25 B.6 Effect of preservatives on the efficacy of glufosinate-ammonium bound to polymer carrier material

In a greenhouse, seeds of the harmful plants ABUTH and BRNSW were sown in humus-containing loam soil, covered with a 0.5 cm layer of the

- same soil and then watered. One day after sowing, a formulation, prepared according to examples 40, 54 and 60, of the herbicide glufosinate-ammonium was broadcast at an application rate of 750 g of active compound per hectare (converted) on the soil. For the entire duration of the experiment, a daytime temperature of 22-24°C and a nighttime temperature of 16-18°C were maintained, a uniform day length of 16 hours being achieved by additional illumination with sodium vapor lamps (about 7000 lux). The relative atmospheric humidity was 60-80%. Visual scoring 35 days after sowing gave the results which showed that the efficacy of the herbicide is enhanced by adding preservatives to glufosinate-ammonium bound to carrier materials, see Table 9.

Table 9 Activity [%] against harmful plants

Treatment with herbicide according for formulation example No.	ABUTH	BRSNW
40 (without preservative)	30	0
54 (with preservative)	55	10
60 (with preservative)	55	70

## Claims:

1. A herbicidal composition, comprising an effective amount of one or more post-emergence herbicides and an amount of a carrier material selected from the group consisting of fullers earth, aerogels, high-molecular-weight polyglycols and polymers based on acrylic acid, methacrylic acid and copolymers thereof.
2. The herbicidal composition as claimed in claim 1, comprising
- a) from 0.15 to 48% by weight of one or more herbicides,
  - b) from 2 to 90% by weight of a carrier material and
  - c) from 0 to 97% by weight of a solvent.
3. The herbicidal composition as claimed in claim 1 or 2, wherein the herbicide is from the group of the herbicides with foliar action.
4. The herbicidal composition as claimed in any of claims 1 to 3, wherein the herbicide is from the group consisting of bilanafos, diquat, glufosinate-ammonium, glyphosate and paraquat.
5. The herbicidal composition as claimed in any of claims 1 to 4, wherein the herbicide is glufosinate-ammonium.
6. The process as claimed in any of claims 1 to 5, wherein the carrier materials are from the group consisting of aerogels, high-molecular-weight polyglycols and polymers based on acrylic acid, methacrylic acid and copolymers thereof.
7. The herbicidal composition as claimed in any of claims 1 to 6, which additionally comprises at least one substance from the group consisting of pre-emergence herbicides, plant growth regulators, fungicides, insecticides, safeners, nutrients, seed dressings and fertilizers.
8. The herbicidal composition as claimed in any of claims 1 to 7, which additionally comprises at least one additive from the group consisting of surfactants, wetting agents, emulsifiers, adjuvants, ammonium salts, preservatives, colorants, antifoams, tackifiers, solvents, buffer systems and UV stabilizers.

9. The herbicidal composition as claimed in claim 8, wherein the additive is from the group consisting of Genapol LRO®, ammonium sulfate and ammonium nitrate.
- 5 10. The use of herbicidal compositions as claimed in any of claims 1 to 9 for controlling undesirable harmful plants, which comprises applying the compositions by the pre-emergence method.
- 10 11. The use of herbicidal compositions as claimed in claim 10, wherein the compositions are used in crops of useful plants which are tolerant to the herbicidally active compounds contained in the compositions.
- 15 12. The use of herbicidal compositions as claimed in claim 11, wherein the compositions are used in crops of genetically modified useful plants.
- 20 13. A method for controlling undesirable harmful plants, wherein the post-emergence herbicides are used in the form of a herbicidal composition as claimed in any of claims 1 to 9 by the pre-emergence method.

**Abstract:**

Herbicidal compositions comprising post-emergence herbicides, carrier materials from the group consisting of fullers earth, aerogels, high-molecular-weight polyglycols and polymers based on acrylic acid, methacrylic acid and copolymers thereof and, if appropriate, further additives, which herbicidal compositions can be used pre-emergence for controlling undesirable harmful plants, are described.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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# **COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY**

As below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below, I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

## **Herbicidal compositions comprising post-emergence herbicides for soil application**

the specification of which

- is attached hereto

- was filed on July 28, 2000 as International Application PCT/EP00/07339

and including all the amendments through the date hereof.

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

## **Prior Foreign Application(s) for which Priority is Claimed:**

Federal Republic of Germany, 19936784.1 of August 9, 1999

And I hereby appoint

William F. Lawrence, Registration No. 28,029, of the firm FROMMER LAWRENCE & HAUG, LLP whose post office address is 745 Fifth Avenue, New York, New York 10151, or their duly appointed associate, my attorneys, with full power of substitution and revocation, to prosecute this application, to make alterations and amendments therein, to file continuation and divisional applications thereof, to receive the Patent, and to transact all business in the Patent and Trademark Office and in the Courts in connection therewith, and specify that all communications about the application are to be directed to the following correspondence address:

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